Optically pumped far-infrared laser lines and frequencies from $^{13}CD_3OH$

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We report 26 new far-infrared laser lines from optically pumped $^{13}CD_3OH$ and the frequencies of 20 of these lines along with 8 other previously reported lines. Most of the new lines were pumped by regular and sequence lines of the 10R branch of a cw CO_2 laser. Nine of the new lines are in the range 32–70 μm, helping to fill the gaps in this wavelength region. To our knowledge, the 32.4-μm laser line is the highest frequency of an optically pumped methanol laser ever measured with the heterodyne technique. The pump offsets for 30 laser lines were also measured (including 20 of the new lines).

1. INTRODUCTION

Methanol and 10 of its isotopic species are the most important far-infrared (FIR) active media, generating more than 2000 laser lines in the range 19–3030 μm.[1,2] Trideuteromethanol-13C ($^{13}CD_3OH$) is one of the most prolific and efficient of the methanol isotopomers, contributing approximately 160 FIR laser lines, with 60% of the wavelengths being shorter than 100 μm. Inguscio *et al.*[3] were the first to investigate this isotopomer as a lasing gas. The importance of this molecule as a laser medium soon became evident, and a great amount of experimental[4,5] and theoretical work[6,7] was undertaken, leading to assignments of laser transitions.

This work reports 26 additional laser lines from $^{13}CD_3OH$, with wavelengths in the range 32.4–469.4 μm. Frequency measurements for 20 of the new as well as 8 previously known $^{13}CD_3OH$ laser lines are also reported.

2. EXPERIMENT

The FIR laser is a metal–dielectric rectangular waveguide cavity, described in detail in Ref. 8. The cavity is pumped by a 1.5-m-long cw CO_2 laser. For the regular and sequence lines of the 10R branch, we used a 171-line/mm grating.[9] For the 9-μm lines we changed to a 163-line/mm grating. Maximum powers are 23 W for the regular lines and 10.5 W for the sequence lines.

The FIR laser frequencies were measured by the method described in Ref. 10. The radiation from two frequency-stabilized CO_2 lasers were mixed with microwave radiation and the unknown FIR laser radiation on a metal–insulator–metal diode. The FIR laser frequency is obtained with the equation

$$\nu_{\mathrm{FIR}} = |n_1\nu_1 - n_2\nu_2| \pm m\nu_{\mu\mathrm{wave}} \pm \nu_{\mathrm{beat}}, \tag{1}$$

where ν_1 and ν_2 are the stabilized CO_2 laser frequencies, $\nu_{\mu\mathrm{wave}}$ is the frequency of the microwave source, ν_{beat} is the beat frequency, and the integers n_1, n_2, and m are harmonic numbers. The intensity of the beat note decreases as the harmonic orders increase, so we usually chose $n_1 = n_2 = m = 1$. When higher frequencies ($\lambda < 60$ μm) were measured, however, this was not always possible. The CO_2 laser frequencies, the microwave frequency, and the harmonic orders are chosen so that ν_{beat} is less than 1.5 GHz. ν_{beat} is amplified and displayed on a spectrum analyzer with a peak-hold feature that records the peak signal as the FIR laser is tuned over its gain curve. The center frequency of the recording is then measured with a marker frequency.

To measure the offset, we mix the pump laser radiation with the radiation from a frequency-stabilized reference laser and measure the beat-note frequency. For the regular band pump line the reference laser operates on the same laser line as the pump. For sequence band pump lines the reference laser operates on a nearby regular band line, and microwave radiation is added to the mix to give a beat note of ~300 MHz.

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Table 1. FIR Laser Lines from $^{13}CD_3OH$

CO_2 Pump[a] Line	λ (μm)	Offset (MHz)	Pressure[b] Pa (mTorr)	Rel. Pol.[c]	Rel. Int.[d]	Pump Power (W)	Ref.
10R(52)	32.427		28 (210)	‖	0.03	12	New
10R(50)′	60.202	+4	117 (880)	‖	5.0	20	New
10R(50)″	81.891	−11	40 (300)	⊥	4.0	20	New
10R(50)‴	51.223[e]	+24	36 (270)	‖	0.5	20	New
10R(46)′	47.583		32 (240)	‖	7.0	23	New
10R(46)″	66.210[f]	+10	20 (150)	‖	5.0	21	New
10R(46)‴	157.679	+42	10 (75)	‖	0.24		5
10R(26)	147.5	−25		⊥			1
10R(24)	64.203	−25	15 (110)	‖	0.2		1
10SR(29)′	469.4	−24	21 (160)	‖	0.1		New
10SR(29)″	219.8	−28	16 (120)	⊥	0.2		New
10SR(27)′	66.667		17 (130)	‖	0.3		New
10SR(27)″	125.071	−13	17 (130)	⊥	0.5	10	New
	160.916	−13	25 (190)	‖	0.5	10	New
10SR(27)‴	70.329	−26	25 (190)	⊥	6.0	10	New
10SR(27)⁗	127.6	+27	17 (130)	‖	0.15	10	New
	234.0	+27	27 (200)	‖	0.05	10	New
10R(12)	82.301		14 (100)	⊥	0.08	16	New
10R(10)	67.302	−2	27 (200)	⊥	0.3	16	4
10R(08)′	97.9		40 (300)	‖	4.0	16	New
10R(08)″	62.499	+30	33 (250)	‖	5.5		1
10R(06)	51.752	+27	28 (210)	‖	2.0		4
10SR(11)	257.628	−15	15 (110)	‖	1.2	8	New
9R(54)	232.351	+16	27 (200)	‖	0.2		New
9R(48)	54.541		45 (350)	‖	0.4		New
9R(34)	82.126	0	33 (250)	‖	2.5		New
9R(32)	55.758	−17	40 (300)	‖	3.0		3
9R(24)	75.275	0	40 (300)	‖	0.6		New
	79.4	0	27 (200)	‖	0.2		New
9R(20)	241.588	+46	27 (200)	‖	0.3		1
9R(18)	52.303	−19	13 (100)	‖	0.2		3, 4
9R(16)	83.146	−45	27 (200)	‖	0.2		New
9R(14)	97.961	+23	33 (250)	‖	2.0		New
9R(10)	65.356	−14	40 (300)	‖	0.5		3
9P(06)′	39.492	−23	40 (300)	‖	0.2		New
9P(06)″	114.556	−4	33 (250)	‖	0.1		New

[a]Symbols ′ and ″ denote different offsets.
[b]Pressure at which each frequency was measured as determined by a pressure gauge (1 Torr = 133.3 Pa).
[c]Rel. Pol., relative polarization.
[d]The 119-μm [9P(36)-pumped] CH_3OH line from this laser has a relative intensity of 10.
[e]Doublet: see Table 2.
[f]Predicted laser line.[6]

3. RESULTS

Table 1 presents the list of the wavelengths of the FIR laser lines observed in this investigation and also other, known lines. The operating pressure, polarization relative to the pump laser, pump offset, CO_2 pump power, and intensity are also listed for most of the lines. Seventeen new laser lines have short wavelengths with $\lambda < 100$ μm. Five of the new laser transitions were pumped by high-J CO_2 laser lines ($J > 50$), and nine others were pumped by sequence lines.

Table 2 shows the results of the frequency measurements. One of the new lines, the 51.2-μm line pumped by 10R(50), is actually a doublet consisting of two lines nearly 4 MHz apart. Also, we were able to measure the frequency of a 32.4-μm laser line at 9.2 THz, which is the highest measured frequency of an optically pumped FIR methanol laser to our knowledge yet observed with the heterodyne technique.[11]

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Table 2. FIR Frequency Measurements of Optically Pumped $^{13}CD_3OH$

λ^a (μm)	CO_2 Pump Line^b	Frequency^c (MHz)	Wave Number^a (cm^{-1})	Ref.
32.427	$10R(52)$	9 245 122.6	308.384096	New
39.492	$9P(06)'$	7 591 236.8	253.216403	New
47.583	$10R(46)'$	6 300 397.4	210.158636	New
51.223	$10R(50)'''$	5 852 724.0	195.225859	New
51.223	$10R(50)'''$	5 852 719.7	195.225715	New
51.752	$10R(06)$	5 792 881.7	193.229734	1
52.303	$9R(18)$	5 731 794.1	191.192071	3, 4
54.581	$9R(48)$	5 492 616.2	183.213955	New
55.758	$9R(32)$	5 376 677.3	179.346650	3
60.202	$10R(50)'$	4 979 742.5	166.106330	New
62.499	$10R(08)$	4 796 751.6	160.002411	1
64.203	$10R(24)$	4 669 448.0	155.756020	1
65.356	$9R(10)$	4 587 083.0	153.008619	3
66.210	$10R(46)''$	4 527 932.5	151.035571	New
66.667	$10SR(27)'$	4 496 880.8	149.999798	New
67.302	$10R(10)$	4 454 427.9	148.583721	1
70.329	$10SR(27)'''$	4 262 687.4	142.187947	New
75.275	$9R(24)$	3 982 631.9	132.846301	New
81.891	$10R(50)''$	3 660 888.4	122.114093	New
82.126	$9R(34)$	3 650 380.0	121.763570	New
82.301	$10R(12)$	3 642 651.6	121.505779	New
83.146	$9R(16)$	3 605 606.5	120.270087	New
97.961	$9R(14)$	3 060 318.9	102.081250	New
114.556	$9P(06)''$	2 616 992.5	87.293474	New
125.071	$10SR(27)''$	2 396 985.6	79.954833	New
160.916	$10SR(27)''$	1 863 038.2	62.144265	New
232.351	$9R(54)$	1 290 254.6	43.038261	New
241.588	$9R(20)$	1 240 925.7	41.392826	1
257.628	$10SR(11)$	1 163 665.7	38.815710	New

^{a}Calculated from the measured frequency with $c = 299\,792\,458$ m/s.
^{b}Symbols $'$ and $''$ denote different offsets.
^{c}Estimated 1σ uncertainty in the reproducibility of the FIR laser frequency $\Delta\nu/\nu = 2 \times 10^{-7}$.

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